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RARE-EARTH, YTTRIUM, AND SCANDIUM ABUNDANCES
IN METEORITIC AND TERRESTRIAL MATTER

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING MAY 15, 1963

Contract NASw-579

National Aeronautics and Space Administration

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May 15, 1963

GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

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This is a preprint of a paper to be submitted for publication
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PREFACE

This Third Quarterly Progress Report of the second year of research for the primary determination of the 14 rare-earth elements (REE) in meteorites covers the contract period from February 15, 1963, to May 15, 1963. In addition to the REE, the abundances of three other important trace elements, yttrium, scandium, and cadmium, have been determined.

During this quarter, the absolute abundances of the REE, Y, Sc, and Cd have been ascertained by neutron-activation analysis in three meteorites and a basalt: a Type I carbonaceous chondrite, Ivuna, obtained from B. H. Mason of the American Museum of Natural History, New York; a calcium-rich achondrite, Stannern, from C. B. Moore of the Niningger Meteorite Collection at Arizona State University; a mesosiderite, Veramin, from E. Olsen of the Chicago Natural History Museum; and a Columbia Plateau basalt (specimen No. I-300) from R. A. McBirney of the University of California, San Diego.

During this quarter, a paper entitled "Abundances of the Fourteen REE, Sc and Y in the Solar System: In Meteoritic, Terrestrial, and Solar Matter" by R. A. Schmitt and R. H. Smith of this Laboratory, in collaboration with Dr. L. Haskin of the University of Wisconsin was presented at the Third Rare-Earth Conference, Clearwater, Florida, April 21-24, 1963. The paper will also be published in the proceedings of the conference.

A paper entitled "Cadmium Abundances in Meteoritic and Terrestrial Matter" by R. A. Schmitt, R. H. Smith, and D. A. Olehy has been submitted for publication in Geochimica et Cosmochimica Acta.

Finally, a paper by R. A. Schmitt, R. H. Smith, and D. A. Olehy, which constitutes the main body of this report and which is entitled "Rare-Earth, Yttrium, and Scandium Abundances in Meteoritic and Terrestrial Matter--II," has been submitted for publication in Geochimica et

Cosmochimica Acta. This last paper discusses REE, Y, and Sc research.

for three quarters under Contract NASw-579 and one-half quarter under

Contract NASw-579.

ABSTRACT

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Abundances and selected isotopic ratios of the 14 rare-earth elements (REE), yttrium; and scandium have been determined by neutron-activation analysis in 13 meteorites and 2 terrestrial specimens: 3 carbonaceous, 2 hypersthene, and 1 bronzitic (troilite phase) chondrite; 2 calcium-rich and 2 nakhlitic achondrites; 1 mesosiderite; 2 pallasites; an Australian eclogite; and a Columbia Plateau basalt. This research is a continuation of recent REE work by Schmitt, et al. (1963a). Absolute abundances of the REE and Sc in 2 Type I carbonaceous chondrites are about 33% less compared with the Type II. Atomic ratios of $Y/10^6Si$ in carbonaceous chondrites remain approximately constant at 4.7 (Type I), 4.1 (Type II), and 4.8 (Type III), and ratios of the REE (La is representative) yielded $La/10^6Si$ at 0.36 (Type I), 0.53 (Type II), and 0.51 (Type III). The REE, Y, and Sc content in the troilite-phase of a chondrite is ~ 0.3 of the content in the entire chondritic matrix. REE and Y abundances in Ca-rich achondrites and nakhlites are ~ 10 and ~ 5 times larger, respectively, than found in ordinary chondrites. No fractionation of the REE and Y distribution was observed in Ca-rich achondrites compared to chondritic REE and Y; fractionation in nakhlites is similar to terrestrial basalts. Fractionation of the REE and Y (Eu is enriched) in the mesosiderite Veramin and in two pallasites (Eu depleted in one pallasite) has been found. The observed fractionation of the REE and Y in Australian eclogite is opposite to fractionation in African eclogite, and fractionation in Columbia Plateau basalt was similar to Kilauea basalt.

A proposed shell-model for a parent chondritic body has been suggested which satisfies the REE, Y, and Sc abundances in all meteoritic phases except Type I carbonaceous and enstatite chondrites, which have been relegated to another parent meteoritic body apart from other chondrites.

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Validity of the shell-model forces increases in chondritic abundances of trace, lithophilic, and nonvolatile elements by a factor of 1.35 to obtain primitive abundances.

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INTRODUCTION

In a recent paper by SCHMITT and SMITH (1962) and SCHMITT, et al. (1963a) (hereafter referred to as REE-I), the abundances and isotopic ratios have been determined for the 14 rare-earth elements (REE) plus Y and Sc by neutron-activation analysis in 19 meteorites and 3 terrestrial specimens. This paper is an extension of that work and reports the determination by neutron activation analysis of the absolute and relative abundances of the 14 REE, Y, and Sc in 13 meteorites and 2 terrestrial specimens. The neutron-activation procedure has been described in REE-I.

Meteoritic and terrestrial specimens were obtained through the courtesy of E. P. Henderson of the Smithsonian Institution (Brenham and Thiel Mountains), J. F. Lovering of the Australian National University (Australian eclogite), B. H. Mason of the American Museum of Natural History (Ivuna, Manbhoom, and Modoc), R. A. McBirney of the University of California, San Diego (Columbia Plateau basalt), C. B. Moore of the Ninninger Meteorite Collection and Arizona State University (Juvinas, Stannern, Nakhla, and Lafayette), B. Nagy of Fordham University (Orgueil), E. Olsen of the Chicago Natural History Museum (Mighei and Veramin). L. T. Silver of the California Institute of Technology kindly separated the troilite of the chondrite Richardton by magnetic separation techniques.

RESULTS AND DISCUSSION

All the abundances obtained for the REE, Y, and Sc in chondritic and nonchondritic meteorites are summarized in Tables 1 and 2. The meteorites are classified according to MASON (1962a, 1962b). The normalized abundances of the REE, Y, and Sc are given in Table 3, where

Table 1

ABSOLUTE ABUNDANCES OF REE, Y, AND Sc IN CHONDRITIC METEORITES AS DETERMINED
BY NEUTRON-ACTIVATION ANALYSIS^a

Isotope Measured	Radio-nuclide	Abundances in 10 ⁻⁶ g/g Sample (ppm)					
		Carbonaceous Chondrites			Hypersthene Chondrites		Bronzitic Chondrite Richardton (Troilite Phase)
		Orgueil	Ivuna	Mighei	Modoc	Manhboom	
La ¹³⁹	40-h La ¹⁴⁰	0.19±0.01	0.19±0.01	0.29±0.01	0.34±0.01	0.31±0.01	0.74±0.04 ^b
Ce ¹⁴⁰	32-d Ce ¹⁴¹	0.66±0.01	0.60±0.01	0.76±0.02	1.42±0.03	0.87±0.01	-----
Ce ¹⁴²	33-h Ce ¹⁴³	0.67±0.01	0.57±0.01	0.71±0.01	1.59±0.05	0.94±0.01	-----
Pr ¹⁴¹	19-h Pr ¹⁴²	0.097±0.001	0.090±0.003	0.13±0.01	0.13±0.01	0.123±0.005	0.058±0.006
Nd ¹⁴⁶	11.1-d Nd ¹⁴⁷	0.44±0.03	0.40±0.02	0.61±0.04	0.63±0.02	0.65±0.005	-----
Sm ¹⁵²	47-h Sm ¹⁵³	0.131±0.002	0.134±0.003	0.20±0.01	0.20±0.01	0.244±0.003	0.080±0.004
Eu ¹⁵¹	9.3-h Eu ^{152m}	0.047±0.002	0.053±0.002	0.078±0.001	0.076±0.002	0.077±0.002	-----
Eu ¹⁵¹	13-y Eu ¹⁵²	0.053±0.002	0.057±0.002	0.077±0.003	0.084±0.005	0.081±0.001	-----
Gd ¹⁵⁸	18-h Gd ¹⁵⁹	-----	0.33±0.05	-----	0.33±0.01	0.26±0.01	-----
Tb ¹⁵⁹	72-d Tb ¹⁶⁰	0.022±0.002		0.047±0.002	0.048±0.001	0.059±0.001	-----
Dy ¹⁶⁴	2.3-h Dy ¹⁶⁵	0.22±0.01	-----	0.36±0.07	0.30±0.02	0.35±0.02	-----
Ho ¹⁶⁵	27-h Ho ¹⁶⁶	0.054±0.001	0.058±0.001	0.076±0.002	0.084±0.002	0.089±0.002	0.034±0.001
Er ¹⁶⁸	9.4-d Er ¹⁶⁹	0.151±0.010	0.146±0.005	0.24±0.02	-----	0.23±0.02	-----
Er ¹⁷⁰	7.5-h Er ¹⁷¹	0.113±0.009	0.151±0.014	0.17±0.01	0.23±0.01	0.25±0.01	-----
Tm ¹⁶⁹	129-d Tm ¹⁷⁰	0.022±0.004	0.022±0.002	0.030±0.004	0.033±0.002	0.036±0.002	-----
Yb ¹⁶⁸	32-d Yb ¹⁶⁹	0.14±0.01	0.103±0.002	0.17±0.01	0.135±0.004	0.21±0.01	-----
Yb ¹⁷⁴	4.2-d Yb ¹⁷⁵	0.17±0.01	0.122±0.003	0.17±0.01	0.164±0.003	0.18±0.01	0.052±0.007
Lu ¹⁷⁶	6.8-d Lu ¹⁷⁷	0.022±0.001	0.024±0.003	0.032±0.005	0.036±0.001	0.033±0.001	0.010±0.001
Y ⁸⁹	64-h Y ⁹⁰	1.44±0.16	1.69±0.19	1.77±0.15	2.07±0.08	1.97±0.11	-----
Sc ⁴⁵	85-d Sc ⁴⁶	5.3±0.2	5.9±0.3	8.6±0.2	8.1±0.2	8.8±0.1	-----

^aValues for elemental abundances were calculated by assuming that terrestrial and meteoritic isotopic ratios were identical for all REE. This assumption has been verified for 5 REE isotopic ratios and particularly for the Yb¹⁶⁸/Yb¹⁷⁴ ratio, which is strongly dependent on nucleosynthetic models. Errors are primarily one standard deviation, due to counting statistics. REE abundances have been rounded to no more than 2 significant numbers after the decimal, zero excluded. The chondritic meteorites have been classified according to MASON (1962a). See also MURTHY and SCHMITT (1963) for additional isotopic analyses of REE.

^bThe high La value in Richardton troilite may possibly be due to contamination because, on a normalized basis, only La is high by a factor of ~6 relative to the other REE.

Table 2
ABSOLUTE ABUNDANCES OF REE, Y, AND Sc IN ACHONDRITIC, MESOSIDERITIC,
AND PALLASITIC METEORITES^a

Isotope Measured	Radio-nuclide	Abundances in 10 ⁻⁶ g/g Sample (ppm)						
		Achondrites (calcium-rich)		Achondrites (Nakhlites)		Mesosiderite	Pallasites	
		Juvinas	Stannern	Nakhla	Lafayette	Veramin (Silicate Phase)	Brenham (Olivine Phase)	Thiel Mts. (Olivine Phase)
La ¹³⁹	40-h La ¹⁴⁰	2.53±0.05	4.89±0.10	1.57±0.05	1.76±0.05	0.29±0.01	-----	0.12±0.01 ^b
Ce ¹⁴⁰	32-d Ce ¹⁴¹	7.29±0.15	12.6±0.2	5.98±0.06	5.50±0.16	0.82±0.02	-----	-----
Ce ¹⁴²	33-h Ce ¹⁴³	7.20±0.15	13.2±0.2	6.40±0.19	5.46±0.13	0.91±0.03	≤0.17±0.01 ^c	0.28±0.03
Pr ¹⁴¹	19-h Pr ¹⁴²	0.98±0.03	1.96±0.04	0.67±0.03	0.80±0.03	0.114±0.005	0.032±0.002	0.022±0.001 ^d
Nd ¹⁴⁶	11, 1-d Nd ¹⁴⁷	5.01±0.12	10.0±0.2	3.18±0.2	3.35±0.06	0.57±0.07	0.16±0.01	0.12±0.01
Sm ¹⁵²	47-h Sm ¹⁵³	1.74±0.03	3.17±0.06	0.73±0.02	0.85±0.02	0.23±0.01	0.112±0.002	0.022±0.001
Eu ¹⁵¹	9, 3-h Eu ^{152m}	0.60±0.02	0.86±0.02	0.20±0.01	0.24±0.01	0.113±0.002	0.018±0.001	0.0031±0.0001
Eu ¹⁵¹	13-y Eu ¹⁵²	0.64±0.02	0.80±0.03	0.19±0.01	0.23±0.01	0.122±0.005	0.017±0.001	-----
Gd ¹⁵⁸	18-h Gd ¹⁵⁹	2.55±0.05	4.47±0.09	0.94±0.03	0.92±0.04	0.44±0.02	-----	0.020±0.003
Tb ¹⁵⁹	72-d Tb ¹⁶⁰	0.40±0.01	0.71±0.02	0.109±0.002	0.120±0.003	0.054±0.004	0.038±0.002	-----
Dy ¹⁶⁴	2, 3-h Dy ¹⁶⁵	3.03±0.10	4.89±0.14	-----	0.89±0.14	0.37±0.01	-----	0.016±0.003
Ho ¹⁶⁵	27-h Ho ¹⁶⁶	0.59±0.01	1.07±0.03	0.140±0.002	0.146±0.004	0.097±0.002	0.051±0.002	0.0034±0.0002
Er ¹⁶⁸	9, 4-d Er ¹⁶⁹	1.70±0.13	2.98±0.15	0.36±0.02	0.47±0.03	0.27±0.03	0.097±0.006	-----
Er ¹⁷⁰	7, 5-h Er ¹⁷¹	1.85±0.03	3.12±0.05	0.32±0.01	0.40±0.01	0.25±0.01	0.091±0.004	-----
Tm ¹⁶⁹	129-d Tm ¹⁷⁰	0.28±0.01	0.47±0.02	0.047±0.003	0.057±0.006	0.041±0.005	0.0096±0.0009	0.0010±0.0002
Yb ¹⁶⁸	32-d Yb ¹⁶⁹	1.37±0.04		0.21±0.01	0.28±0.02		-----	-----
Yb ¹⁷⁴	4, 2-d Yb ¹⁷⁵	1.37±0.04	2.36±0.08	0.24±0.01	0.36±0.01	0.23±0.01	0.043±0.001	0.0086±0.0005
Lu ¹⁷⁶	8-d Lu ¹⁷⁷	0.231±0.004	0.38±0.01	0.044±0.001	0.051±0.002	0.044±0.002	0.0059±0.0004	0.0017±0.0002
Y ⁸⁹	64-h Y ⁹⁰	17.1±0.3	28±0.5	3.17±0.15	4.4±0.2	2.47±0.17	1.57±0.19	-----
Sc ⁴⁵	85-d Sc ⁴⁶	28±0.4	31±0.6	54±1.1	78±1.5	16.2±0.3	0.79±0.03	1.15±0.02

^aValues for elemental abundances were calculated by assuming that terrestrial and meteoritic isotopic ratios were identical for all REE. This assumption has been verified for 5 REE isotopic ratios and particularly for the Yb¹⁶⁸/Yb¹⁷⁴ ratio, which is strongly dependent on nucleosynthetic models. Errors are primarily one standard deviation, due to counting statistics. REE abundances have been rounded to no more than 2 significant numbers after the decimal, zero excluded. The chondritic meteorites have been classified according to MASON (1962a). See also MURTHY and SCHMITT (1963) for additional isotopic analyses of REE.

^bThe La value for another olivine portion in Thiel Mountains was 0.022±0.001 ppm.

^cThe upper limit to Ce was calculated because of the presence of Ce¹⁴⁴ gamma rays that were possibly due to neutron-induced fission of uranium in olivine.

^dThe value given for Pr was obtained from the first neutron-activation analysis of another olivine portion of Thiel Mountains. In the 2 Thiel Mountains analyses the La/Pr ratios were 5.5 and 1.8, respectively. The source of the discrepancy is unknown, but the value given for Pr agrees well with the other REE when normalized to that of the chondrites.

Table 3

NORMALIZED ABUNDANCES OF REE, Y, AND Sc IN CHONDRITIC, ACHONDRITIC, MESOSIDERITIC, AND PALLASITIC METEORITES

Element	Chondrites						Achondrites (calcium-rich)		Achondrites (Nakhlites)		Meso- siderite	Pallasites (Olivine phase)		Average in
	Orgueil (Carb.)	Ivuna (Carb.)	Mighei (Carb.)	Modoc (Hyp.)	Manbhoom (Hyp.)	Richardton (Bron. Troilite)	Juvinas	Stannern	Nakhla	Lafayette	Veramin	Brenham	Thiel Mts.	17 chondrites ^a
La	1.00	1.00	1.00	1.00	1.00	6.1 ^b	1.00	1.00	1.00	1.00	1.00	(b)	1.00	1.00
Ce	3.50	3.08	2.62	4.40	2.81	-----	2.86	2.64	3.94	3.12	2.98	≤1.00	2.33	2.80±0.23 ^c
Pr	0.51	0.47	0.45	0.38	0.40	0.48	0.39	0.40	0.43	0.45	0.39	0.19	0.18	0.41±0.005
Nd	2.31	2.10	2.10	1.85	2.10	-----	1.98	2.05	2.02	1.90	1.97	0.91	1.01	1.88±0.16
Sm	0.69	0.71	0.69	0.59	0.78	0.66	0.69	0.65	0.47	0.48	0.79	0.66	0.18	0.66±0.05
Eu	0.26	0.29	0.27	0.24	0.25	-----	0.25	0.176	0.124	0.134	0.40	0.13	0.026	0.24±0.03
Gd	----	0.174	----	0.97	0.84	-----	1.01	0.92	0.60	0.52	1.52	----	0.17	0.97±0.11 ^d
Tb	0.12		0.16	0.14	0.19	-----	0.158	0.145	0.069	0.068	0.19	0.22	-----	0.16±0.02
Dy	1.16	----	1.24	0.88	1.13	-----	1.20	1.00	----	0.51	1.28	----	0.13	0.99±0.13
Ho	0.28	0.31	0.26	0.25	0.29	0.28	0.23	0.22	0.089	0.083	0.34	0.30	0.027	0.24±0.03
Er	0.68	0.79	0.71	0.68	0.81	-----	0.70	0.63	0.216	0.247	0.90	0.55	-----	0.69±0.06
Tm	0.116	0.116	0.103	0.097	0.116	-----	0.111	0.096	0.030	0.032	0.141	0.057	0.0083	0.103±0.008 ^e
Yb	0.82	0.60	0.52	0.45	0.63	0.43	0.54	0.48	0.143	0.182	0.79	0.25	0.072	0.56±0.05 ^f
Lu	0.116	0.126	0.11	0.106	0.11	0.082	0.091	0.078	0.028	0.028	0.152	0.035	0.014	0.105±0.012
Y	7.6	8.9	6.1	6.1	6.4	-----	6.8	5.7	2.02	2.50	8.5	9.2	-----	6.5±0.5 ^g
Sc	28	31	30	24	28	-----	11.0	6.2	34	44	56	4.7	9.6	27±3 ^h

^aAverage values include 12 chondrites given in Table 3 of SCHMITT, *et al.* (1963a) and 5 chondrites of this Table. Average abundance of La = 0.30 ppm in 17 chondrites.

^bAbundances normalized to Sm at 0.66.

^cAbundances of Allegan, Richardton, Holbrook, Modoc, and Orgueil excluded.

^dAbundance of Ivuna excluded.

^eAbundance of Forest City excluded.

^fAbundance of Orgueil excluded.

^gAbundance of Ivuna excluded; average of 11 chondrites given.

^hAbundance of Holbrook excluded; average of 11 chondrites given.

for comparative purposes, the average normalized REE, Y, and Sc abundances for 17 chondritic meteorites are given in the last column. It may be remarked parenthetically that the contribution to Ce^{141} and Ce^{143} from the neutron-induced fission of the uranium and thorium present in these chondrites is only about 1% to 2%, and therefore negligible. The constancy of the meteoritic $\text{Ce}^{141}/\text{Ce}^{143}$ ratio relative to a pure Ce standard and the absence of enhanced Nd abundance via the 11.1-d Nd^{147} radioisotope (also a possible fission product) rules out significant neutron-fission contribution to the REE abundances.

GENERAL ABUNDANCES IN CHONDRITIC METEORITES

1. Within experimental error, most of the normalized REE and Sc abundances in Orgueil and Ivuna agree with the normalized average in 17 chondrites. On an absolute basis, the concentrations of the REE and Sc in Orgueil and Ivuna are rather low--i. e., the abundance of La, a representative REE, is 0.19 ppm, whereas the average La abundance in 17 chondrites is 0.30 ppm.

An atomic ratio of $\text{La}/10^6\text{Si}$ is calculated at 0.36 ± 0.01 for Type I Orgueil and Ivuna, and 0.53 ± 0.07 is found in Type II carbonaceous chondrites Mighei and Murray (see REE-I and Table 1 of this paper). $\text{La}/10^6\text{Si}$ values of 0.51 ± 0.08 , 0.38 ± 0.07 , 0.38 ± 0.03 , and 0.24 ± 0.04 were obtained for pigeonitic, hypersthénic, bronzitic, and enstatitic chondrites, respectively. These atomic ratios indicate that the enstatitic chondrites deviate significantly from the other four chondritic categories--i. e., the contents of the trace REE and Sc appear to be markedly depleted relative to Si in enstatitic chondrites. On the basis of weight (H_2O and C excluded), the abundances of the REE and Sc in Type I carbonaceous and enstatitic chondrites agree well (these are discussed in a later section, as are the abundances of Y).

The low atomic ratio of the trace element La at 0.36 for Type I carbonaceous chondrites compared with higher values in other carbonaceous classes may result from some inhomogeneity of chondritic matter during

the initial accretion of primitive matter (WOOD, 1961) or as a result of subsequent physiochemical processes. Since abundances in the percentile range of such elements as Al, S, Na, and Ca fluctuate appreciably in the various carbonaceous classes (WIIK, 1956), variations in trace-element abundances within the carbonaceous chondritic classes should not be unexpected.

2. The normalized abundances of Ce and Yb appear to be enhanced in Orgueil relative to the average in 17 chondrites. With Tm and Lu, which are adjacent to Yb, apparently unaffected in normalized abundances, no account can be offered for the apparent discrepancy of Yb. One measure of the meteoritic oxidation history may be found in the comparisons of the Ce/La ratios. A high Ce/La ratio of 3.50 in Orgueil may be indicative of an above-average oxidation environment during mineral formation compared to the environment of Types I and II carbonaceous chondrites Ivuna, Murray, and Mighei, which have lower Ce/La ratios of 3.08, 2.69, and 2.55, respectively (see REE-I). The element Ce, upon oxidation to the +4 state, may crystallize in lattices different from the common trivalent REE minerals. Since such processes are ordinarily restricted to highly mobile and diffusive conditions common to the liquid or semiliquid state at high temperatures, it is highly probable that any organisms, if present, would be destroyed during this particular phase of chondritic history. (The possible existence of fossilized organisms in Type I carbonaceous chondrites has been discussed at some length in "Life Forms in Meteorites," (1962)).

The high Ce/La ratio in Orgueil may be interpreted in the light of the recent discussion by DUFRESNE and ANDERS (1962) on the chemical evolution of the carbonaceous chondrites. Evidence has been presented which suggests that these carbonaceous meteorites and the associated minerals have been exposed to the chemical action of water. If the REE had precipitated in separate minerals, such as carbonates, and had been exposed to an oxidizing alkaline medium for oxidation of Ce^{+3} to Ce^{+4} ($E^0 = -1.61$), percolation of water through such a mineral system would

have preferentially dissolved the trivalent REE, e. g., the solubility product of $\text{Ce}(\text{OH})_3$ at 25°C is $\sim 10^{-48}$, whereas the $\text{REE}(\text{OH})_3$ solubility products vary uniformly from $\sim 10^{-19}$ to $\sim 10^{-24}$ for $\text{La}(\text{OH})_3$ through $\text{Lu}(\text{OH})_3$. (Dolomite, a carbonate, has been found in both Orgueil and Ivuna.) The observed $\text{La}/10^6\text{Si}$ atomic ratio of 0.36 could be explained if $\sim 1/3$ of the trivalent REE initially present (in amounts corresponding to the average of $0.53 \pm 0.07 \text{ La}/10^6\text{Si}$ in Type II Mighei and Murray) were dissolved. Retention of all Ce^{+4} in the REE mineral upon water erosion would yield a Ce/La ratio of 3.5, which agrees with the observed value of 3.50. If the REE are scattered homogeneously through the common silicate minerals, one third of these silicate minerals must necessarily be dissolved.

Dissolution of small REE minerals with large surface-to-volume ratios would require a minimum of percolation compared to dissolution of larger meteoritic minerals; consequently, other trace elements, if present homogeneously in the larger crystal lattices, would be comparatively unaffected, because larger quantities of water would be required to dissolve these phases.

The percolation mechanism given above may, however, be invalid for the following reasons. DUFRESNE and ANDERS (1962) have noted that the magnesium abundances in all carbonaceous chondrites are relatively constant and that large quantities of magnesium sulfate have been synthesized on the spot and not transported great distances within the Type I carbonaceous chondrites. A percolation mechanism most certainly would have removed appreciable fractions of the very soluble magnesium sulfate while transporting dissolved minerals containing the REE. No such transport of magnesium sulfate has been observed. On the other hand, small REE minerals could be dissolved and transported in quantities of water that would also move comparatively small amounts of the magnesium sulfate. Determination of other trace lithophilic elements having dissimilar chemical properties compared with the REE may help clarify this problem.

The analyzed samples of Orgueil and Ivuna were only 1.7 and 1.0 g, respectively. However, in all other small quantities of carbonaceous and

ordinary chondrites, analyzed by other investigators, the abundances of selected REE agreed within experimental error with those of this work and REE I.

Another serious difficulty with a percolation mechanism is that the high potential that is required for $\text{Ce}^{+3}-\text{Ce}^{+4}$ oxidation is not consistent with the presence of reduced species (e.g., FeS and hydrocarbons) in Type I carbonaceous chondrites.

In summary, the conditions for the explanation of the low $\text{La}/10^6\text{Si}$ ratio of 0.36 in Type I carbonaceous chondrites by a percolation mechanism seem to be too restrictive. In view of the absence of magnesium sulfate transport and of the high potential of Ce oxidation, the low REE abundance with respect to silicon and the different Ce/La ratios in Type I carbonaceous chondrites indicate fractionation of the whole REE series, and of Ce with respect to the other trivalent REE. This most probably occurred before the accretion of the carbonaceous chondrites in a hot portion of the solar nebula (WOOD, 1962).

REED, et al. (1960), found by neutron-activation analysis a lower uranium concentration of 0.008 ppm in Orgueil compared with 0.016 ppm in Mighei, and GOLES and ANDERS (1962) found 0.015 ppm and 0.020 ppm in Mighei and Murray, respectively. If the percolation mechanism is correct, uranium must be relegated to small soluble phases in Orgueil. Abundances of barium, another trace lithophilic and nonvolatile element, in Orgueil were similar to Mighei at 2.4 and 2.5 ppm, respectively (REED, et al., 1960). On the other hand, PINSON, et al. (1953), using spectrochemical techniques, reported <1, <1, <1, and 4 ppm for Ba, Sr, Zr, and Sc abundances in Orgueil, respectively. The Sc content of 4 ppm in Orgueil agrees within ~30% with the Sc value of 5.3 ppm reported in this work. The hiatus between the neutron activation and spectrochemical data for the trace elements Ba, Sr, and Zr allows no clear comparisons between this data and the REE, Y, and Sc data in Orgueil.

An examination of all the Ce/La ratios in REE-I and Table 3 of this paper reveals essentially 3 arbitrary groups of Ce/La values with no clear

distinction among the 5 chondritic categories. Ce/La ratios of 2.30, 2.52, 2.55, 2.69, 2.64, 2.81, and 2.81 (2.30 to 2.81 group) were obtained for 2 carbonaceous pigeonitic, 2 Type II carbonaceous, 2 enstatitic, and 1 hypersthene chondrite, respectively; Ce/La ratios of 3.08, 3.50, 3.22, 3.04, 3.06 (3.04 to 3.50 group) exist for 2 Type I carbonaceous, 1 enstatitic, 1 bronzitic, and 1 hypersthene chondrite, respectively. Finally, the 2 hypersthene chondrites Holbrook and Modoc, which constitute the last group, have especially high Ce/La ratios at 4.71 and 4.40, respectively. Unreliable ratios of Ce/La for Allegan and Richardton have been neglected (these two meteorites were the first analyzed). Such an apparent random distribution in Ce/La ratios complicates interpretation of the physiochemical histories of the individual meteorites.

3. Absolute and relative abundances of the REE, Y, and Sc in Mighei and Modoc agree with the average values in 17 chondrites. The single exception, Ce in Modoc, has been noted above.

It is noted by ZAOZERSKII and PATKIN (1959) that Ce may be easily oxidized to the tetravalent state by atmospheric oxygen under increased pH and temperature conditions. If the REE are introduced into the crystallizing matrix of the chondrite, with enhancement of the Ce^{+4} abundance, it is conceivable that the abundances of other tetravalent ions, such as Ti, Zr, Hf, and Th, may be increased in these hypersthene chondrites. Also, oxidation of the two hypersthene chondrites Modoc and Holbrook is not inconsistent with the observation that the FeO content in hypersthene chondrites is considerably larger (because of more oxidation) than in bronzitic chondrites.

Absolute values in Modoc of 0.080 ± 0.001 and 9.2 ppm for Eu and Sc, respectively, as determined by BATE, et al. (1960), by neutron-activation analysis, agree with the Eu and Sc values of this work-- 0.080 ± 0.004 and 8.1 ± 0.2 ppm, respectively. An estimated 3% error on the Sc value of 9.2 by BATE, et al., brings the Sc agreement within the 95% confidence level. Such close agreement again underscores the homogeneity of chondritic meteorites.

ABUNDANCES OF REE, Y, AND Sc IN AMPHOTERIC CHONDRITE MANBHOOM

Absolute and relative REE, Y, and Sc abundances in the amphoteric chondrite Manbhoom agree well with those found in the ordinary chondrites. KVASHA (1958) and MASON (1962a) have categorized this chondrite, which contains very little free metal, as an olivine hypersthene. Furthermore, the abundance in Manbhoom of another critical trace element, Cd, which is a chalcophilic element, agrees with Cd abundances of hypersthenic and bronzitic chondrites (SCHMITT, et al., 1963b). With a "normal" Ce/La ratio of 2.81, the hypersthenic chondrite Manbhoom is, with regard to oxidation history, more closely related to Kyushu than to Modoc and Holbrook.

ABUNDANCES OF SELECTED REE IN TROILITE PHASE

Absolute concentrations of the five REE Pr, Sm, Ho, Yb, and Lu in the troilite (FeS) phase of the bronzitic chondrite Richardton are $\sim 1/3$ to $1/2$ as large as they are in the whole meteorite. The concentration of La at 0.74 ppm seems to be twice as large as the average La value in chondritic matter and therefore may be due to contamination. In view of the lithophilic character of the REE, such a depletion of 2 or more was anticipated in the troilite phase.

YTTRIUM ABUNDANCES IN CHONDRITES

Previous work (REE-I) reported the absolute abundances of Y in seven chondrites to be from 1.02 to 2.42 ppm, averaging 1.9 ± 0.4 ppm, with a $Y/10^6Si$ atomic ratio of 3.6 ± 0.7 . Including the five chondritic Y values reported in this work, the average Y abundance in 12 chondrites is 1.8 ± 0.4 ppm, which corresponds to $3.4 \pm 0.7 Y/10^6Si$. This value is much lower than the SUESS-UREY (1956) atomic value of $8.9 Y/10^6Si$.

Again, for the element Y, the atomic abundance (based on more meteorites than given in REE-I) expressed in $Y/10^6Si$ decreases regularly from the carbonaceous to the enstatitic meteorites: 4.7 ± 0.4 in 2 Type I

carbonaceous: 4.1 in 1 Type II carbonaceous; 4.8 ± 0.1 in 2 Type III carbonaceous pigeonitic; 3.4 ± 0.2 in 4 bronzitic and hypersthene; and 2.3 ± 0.5 in 2 enstatitic chondrites. Clearly, there is no appreciable fractionation of Y among the carbonaceous chondrites as has been observed for the REE among the carbonaceous chondrites (see above, "General Abundances in Chondritic Meteorites," observation 1), where $\text{La}/10^6\text{Si} = 0.36 \pm 0.01$ in 2 Type I carbonaceous, 0.53 ± 0.07 in Table II carbonaceous, and 0.51 ± 0.08 in 2 carbonaceous pigeonitic chondrites. It seems highly improbable that any systematic error could have been introduced in the REE and Y analyses to give this striking fractionation difference between the REE and Y in the carbonaceous chondrites. Chronologically, the analysis of Orgueil followed that of the amphoteric chondrite Manbhoom. Six other meteoritic and terrestrial specimens were analyzed after Orgueil and before the Ivuna analysis: these six--in chronological order--were the eclogite Juvinas, Lafayette, Stannern, Columbia basalt, and Veramin. Since the geochemical properties of Y are very similar to those of the heavy REE (the ionic radius of Y is identical to that of Dy), it was predicted that the Y/REE or Y/La ratio should remain essentially constant for the various chondritic classes. In general, this has been confirmed, with Y/La ratios of 7.7 for Type II carbonaceous, 9.4 for carbonaceous pigeonitic, 9.0 for bronzitic and hypersthene, and 9.6 for enstatitic chondrites. The ratio of 7.7 is based on only one Y value. However, within the Type I carbonaceous class, where $\text{Y/La} = 13.1$, fractionation of $\sim 50\%$ has occurred between Y and the REE.

GREENLAND (1963) has reported fractionation of Zn and Ge within the carbonaceous chondritic group. The chalcophilic properties responsible for the Zn and Ge fractionation, may hardly be attributed to the REE and Y. Invoking a percolation mechanism for the depletion of the REE would also fractionate Y to the same degree as the heavy REE (there was no fractionation within the REE group as a whole, Ce excepted), since the solubility products of Y(OH)_3 and the heavy REE(OH)_3 are of the same magnitude. No simple differentiation mechanism is known to the authors for separating

Y from the REE by $\sim 50\%$ without associated internal REE fractionation. In fact, all observed fractionations of the REE and Y in achondrites reported in this work and in REE-I have differentiated Y and Dy (same ionic radii) to practically the same degree relative to Y and Dy in all chondritic classes except the Type I carbonaceous. The absence of a plausible geochemical explanation for the REE fractionation in the carbonaceous chondrites suggests that the Type I carbonaceous chondrites may possibly have originated from a different nucleosynthetic source than the other chondrites. Changes in normalized REE abundances and in sensitive isotopic ratios serve as checks, since Y and the light REE are synthesized (BURBIDGE, et al., 1957, and CLAYTON, et al., 1961) predominantly by the s-process and the heavy REE principally by the r-process. With the exception of Ce, most of the REE abundances and isotopic ratios agree within experimental error, whereas the elemental abundances of Gd in Ivuna and Yb in Orgueil are high and unexplainable at the moment.

Since Yb^{168} is produced via the p-process and Yb^{174} via the s- and r-processes, the agreement within the 95% confidence level of the ratio $\text{Yb}^{168}/\text{Yb}^{174}$ in Orgueil and Ivuna would rule out appreciable synthesis from another source. Also, since Yb^{168} and Yb^{174} have thermal-neutron cross sections of 11,000 and 60 barns, respectively, the 95% confidence-level agreement between $\text{Yb}^{168}/\text{Yb}^{174}$ in Type I meteoritic and terrestrial matter rules out any significant differences in thermal-neutron irradiation exposures that may have occurred for meteoritic and terrestrial matter (FOWLER, et al., 1962). In addition, the near identity of the $\text{Ce}^{140}/\text{Ce}^{142}$ isotopic ratios in Orgueil and Ivuna (see Table 1) to corresponding ratios in other meteoritic and terrestrial matter does not indicate different nucleosynthetic sources. Ce^{140} and Ce^{142} are synthesized via the s- and r-processes, respectively (BURBIDGE, et al., 1957, and CLAYTON, et al., 1961).

For four meteorites Forksville, Holbrook, Pasamonte, and Murray (a Type II carbonaceous chondrite), MURTHY and SCHMITT (1963) have

found indistinguishable isotopic ratios to $\pm 0.5\%$ for the sensitive REE, Sm, Eu, and Gd (some of whose isotopes have extremely high thermal-neutron cross sections) compared to standard terrestrial matter. Their technique, a combination of neutron-activation and mass-spectrometric methods, could check the possibility that Type I carbonaceous chondrites originated from a different nucleosynthetic source or were exposed to different thermal-neutron exposures than were other chondrites and terrestrial matter.

ABUNDANCES OF REE, Y, AND Sc IN Ca-RICH ACHONDRITES

Absolute abundances of the REE and Sc in the eucritic Ca-rich achondrites Juvinas and Stannern (La = 2.53 ppm and 4.89 ppm, respectively) agree well with two previous analyses of Ca-rich achondrites, eucritic Pasamonte (La = 3.21 ppm) and howarditic Nuevo Laredo (La = 4.03 ppm) (see REE-I). More significant is the observation that the normalized abundances of the REE in all four of these Ca-rich achondrites (and also Y in two achondrites) are identical to the REE in chondrites. On the average, the absolute REE and Y abundances in Ca-rich achondrites are ~ 12 times more abundant than in chondrites, and the Sc has been enriched ~ 4 times in the four Ca-rich achondrites relative to the Sc in chondrites. If these achondrites have been derived from basic chondritic matter, an equivalent observation is that the chondritic matter has retained ~ 3 times more Sc compared to its retention of the REE and Y. This will be dealt with in the last section.

A plot (Fig. 1) of the ratios of the normalized weight percent of the REE and Y in achondrites to the normalized REE and Y in chondrites clearly shows nonfractionation of the REE and Y in Ca-rich achondrites compared to Ca-poor achondrites. However, Sc has been fractionated in all achondrites to significant degrees (Norton County excepted) compared to the extrapolated values. See REE-I for a suggested explanation based on the geochemical similarity between scandium, magnesium, and divalent iron (GOLDSCHMIDT, 1954).

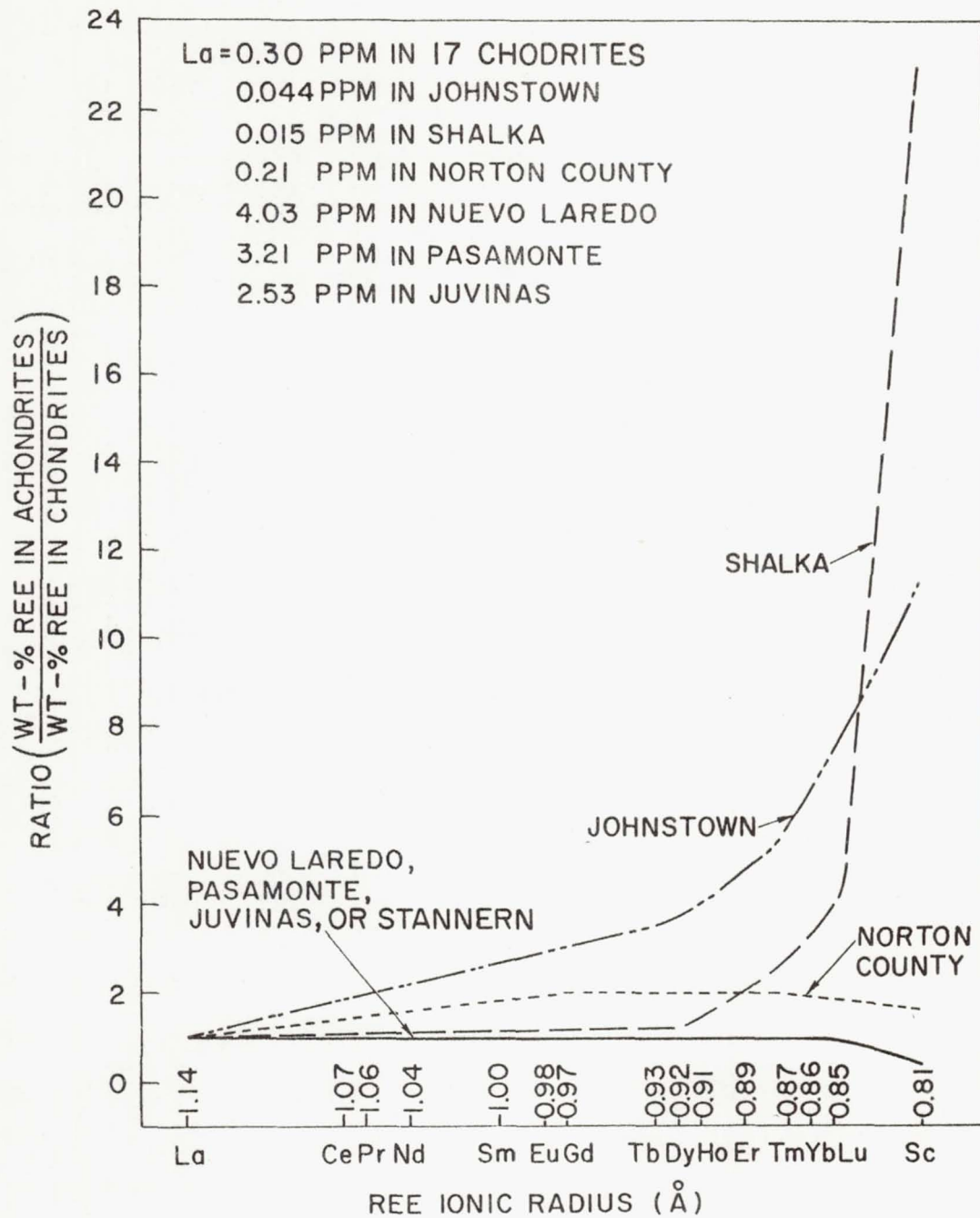


Fig. 1--Ratios of normalized weight percent of the REE in the "chondrite-like;" or Ca-poor, achondrites Johnstown, Shalka, and Norton County and in the "basalt-type," or Ca-rich, achondrites Neuvo Laredo, Pasamonte, Juvinas, and Stannern to those in 17 chondrites as a function of trivalent ionic radius

FRACTIONATION OF REE, Y, AND Sc IN NAKHLITIC METEORITES

Differences in the abundances of the REE, Y, and Sc in the two nakhlitic meteorites relative to their abundances in ordinary chondrites have been recently reported by SCHMITT and SMITH (1963). In short, the similarity in REE and Y fractionation in these nakhlitic meteorites relative to the REE and Y in terrestrial basalts (see Fig. 2) suggested that these two meteorites originated from terrestrial-like volcanic action on the parent meteoritic body.

FRACTIONATION OF REE, Y, AND Sc IN MESOSIDERITIC METEORITES

Uniform fractionation (see Fig. 3) of the REE and Y relative to chondrites has been found in the silicate phase in Veramin, a mesosiderite, and an observed "fall." This contrasts sharply with nonfractionation of the REE and Y in the mesosiderite Estherville (see REE-I). On an absolute basis, the REE and Y were ~ 4 times more abundant in Estherville than in chondrites versus approximately the same REE and Y abundances in Veramin compared with chondrites. Note in Fig. 3 that Lu and Yb, the heaviest REE, are enriched by ~ 1.4 . In the derivation of mesosiderites from some common siliceous matter, Sc has apparently been enriched to about the same degree (to a factor of ~ 2) in both Estherville and Veramin, since the Sc abundance in these two mesosiderites is 17.8 ppm and 16.2 ppm, respectively.

The element Eu has been enriched in Veramin by ~ 1.4 relative to Eu in chondrites (see Fig. 3). This is the first meteorite among 32 analyzed in which Eu has been enriched. Only in the Ca-poor achondrites and pallasites has Eu been depleted--i. e., in Norton County by ~ 4.2 , in Johnstown by ~ 3.3 , and in Brenham by ~ 1.8 (see below). This suggests

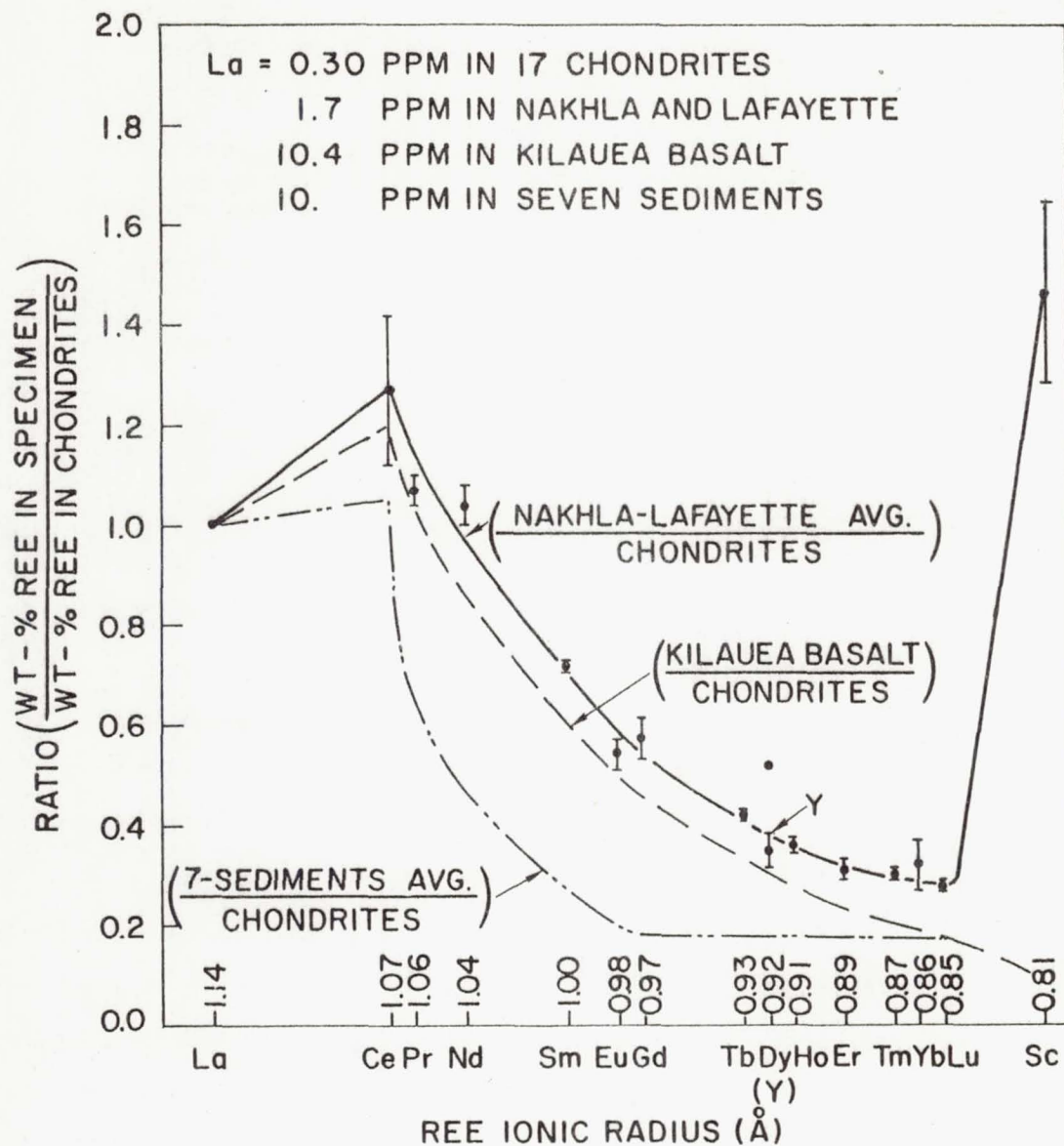


Fig. 2--Ratios of the average normalized weight percent of the REE in Nakhla and Lafayette to those in 17 chondrites: mean deviations are indicated for each REE except Dy (determined only in Lafayette). Comparative curves of ratios for REE in Kilauea Iki-22 basalt by SCHMITT, *et al.* (1963), and for the REE in average of 7 sediments by HASKIN and GEHL (1962) are included. Ratios are plotted as a function of trivalent ionic radius

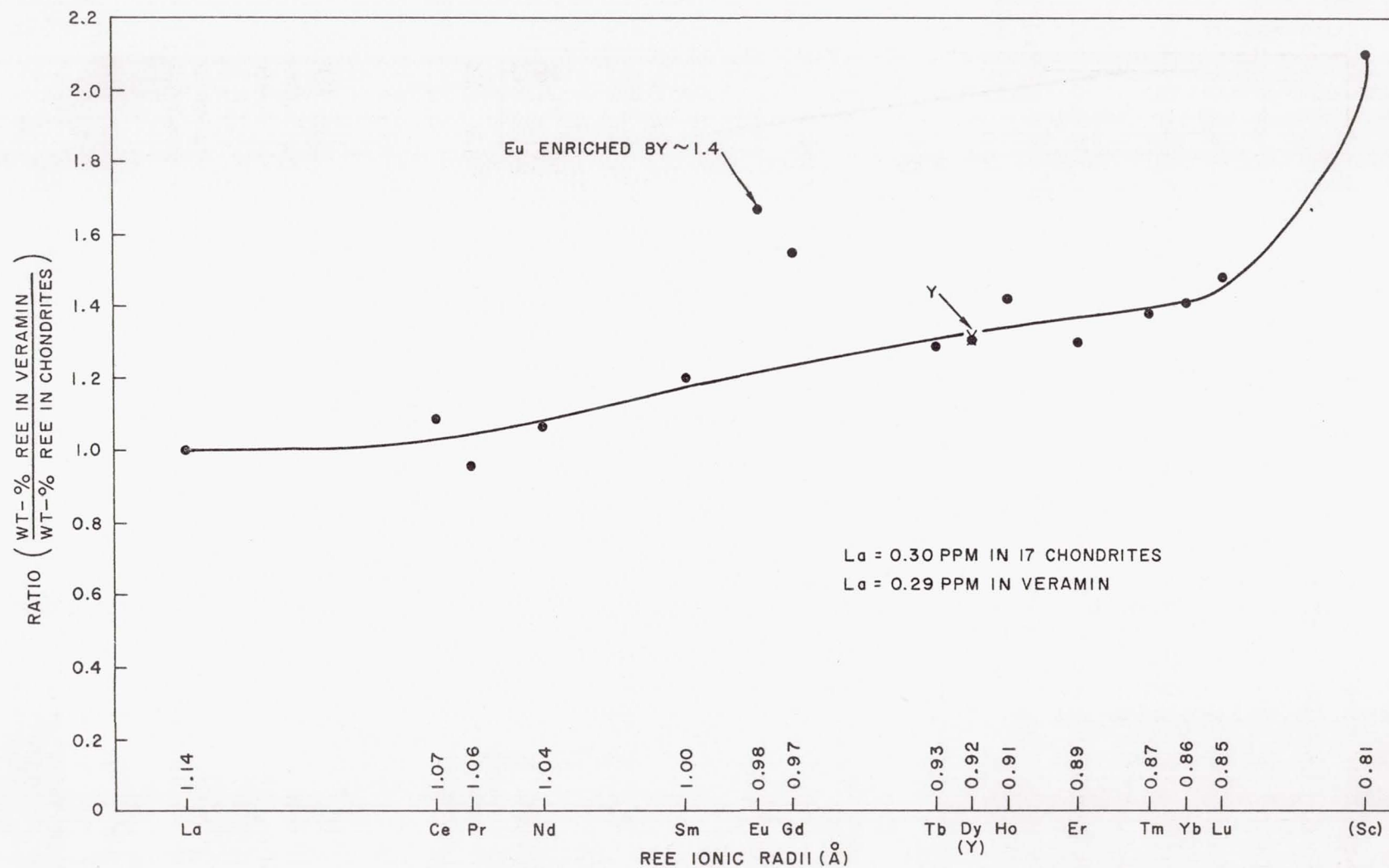


Fig. 3--Ratios of normalized weight percent of REE, Y, and Sc in the mesosiderite Veramin to those in 17 chondrites as a function of trivalent ionic radius

that some of the mesosiderites may be the complements of Ca-poor achondrites and pallasites. The availability of a comparatively stable divalent oxidation state for Eu relative to the adjacent medium weight REE in the trivalent state, may readily account for the enhancement and complementary depletion of the Eu during differentiation under appropriate reduction conditions (see REE-I).

No great confidence should be placed on the apparent Gd enrichment of ~ 1.24 in Veramin relative to chondrites. Because of possible overlapping of Eu and Gd during the ion-exchange elution, the Gd abundance would actually be lower.

FRACTIONATION OF REE, Y, AND Sc IN PALLASITIC METEORITES

Absolute abundances of the REE, Y, and Sc in the olivine phase of the two pallasites Brenham and Thiel Mountains were about 0.05 to 0.5 times the abundances in chondritic meteorites. In both pallasites (both were "finds"), the REE, Y, and Sc have been fractionated differently (see Figs. 4 and 5). The question of terrestrial contamination may not be entirely ruled out. E. P. Henderson selected olivine crystals with the least (if any) contamination. Fractionation of the REE in Thiel Mountains strongly resembles terrestrial fractionation of REE found in Kilauea Iki-22 basalt and peridotite and in seven terrestrial sediments by Haskin and Gehl (1962) (see REE-I); therefore, terrestrial contamination of the Thiel Mountains may not be completely ruled out. In Brenham, the fractionation of the REE, Y, and Sc is completely different from terrestrial basalts, peridotites, etc. Certain rare-earth minerals, such as samarskite $[(\text{Gd}, \text{U}, \text{Fe})(\text{Nb}, \text{Ta})_2\text{O}_6]$, enrich the medium-weight REE by factors of ~ 100 to ~ 40 with respect to La and Lu, respectively (SEMENOV and BARINSKII, 1958). Enrichment factors have been calculated relative to normalized REE abundances in chondritic meteorites calculated by CORYELL, *et al.* (1962). In samarskite the element Eu was depleted by a factor of ~ 30

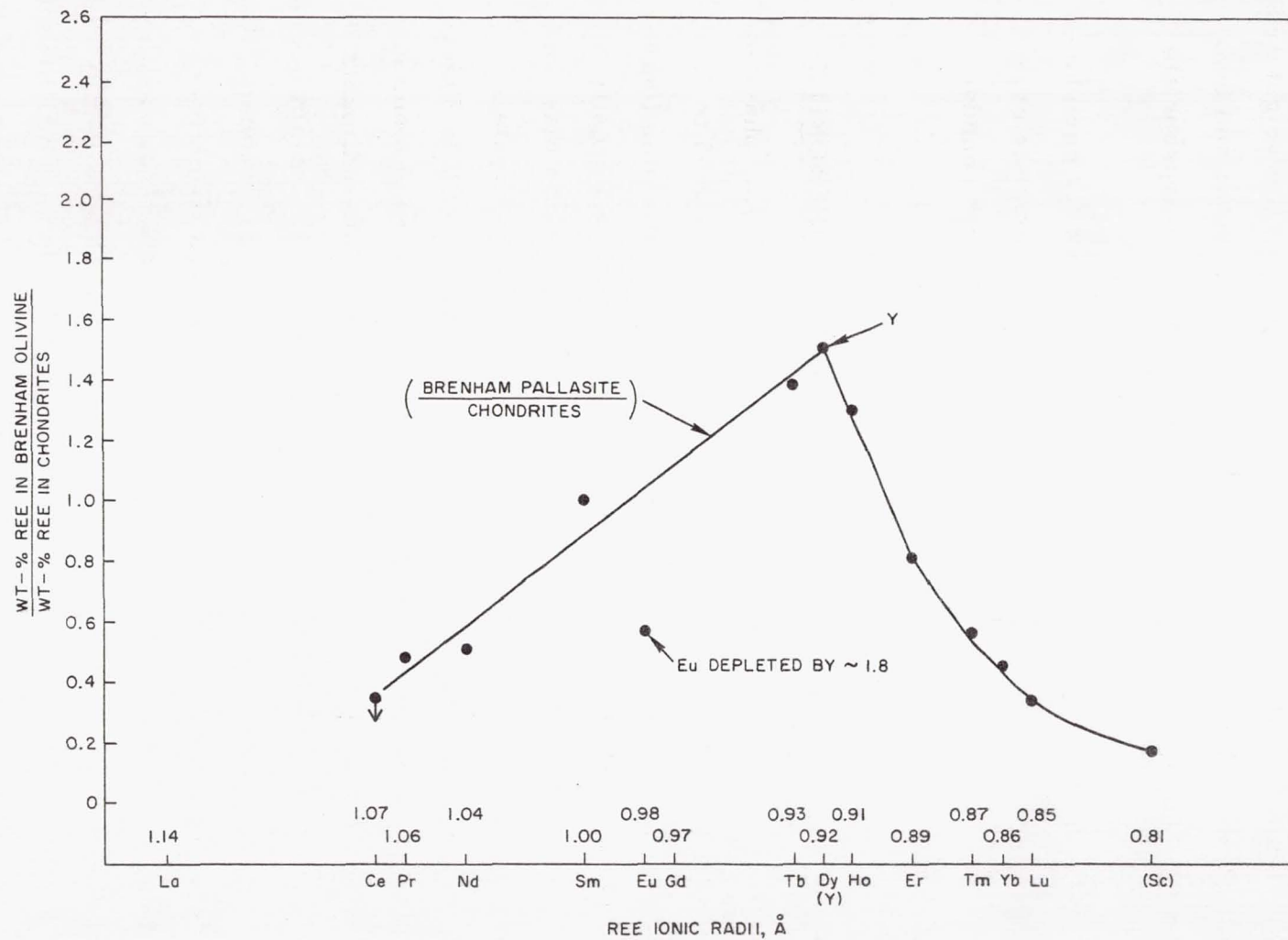


Fig. 4--Ratios of the normalized (to Sm at 0.66) weight-% REE in Brenham pallasite (olivine phase) to the average normalized weight-% REE in 17 chondrites as a function of ionic radii in Å units of trivalent REE

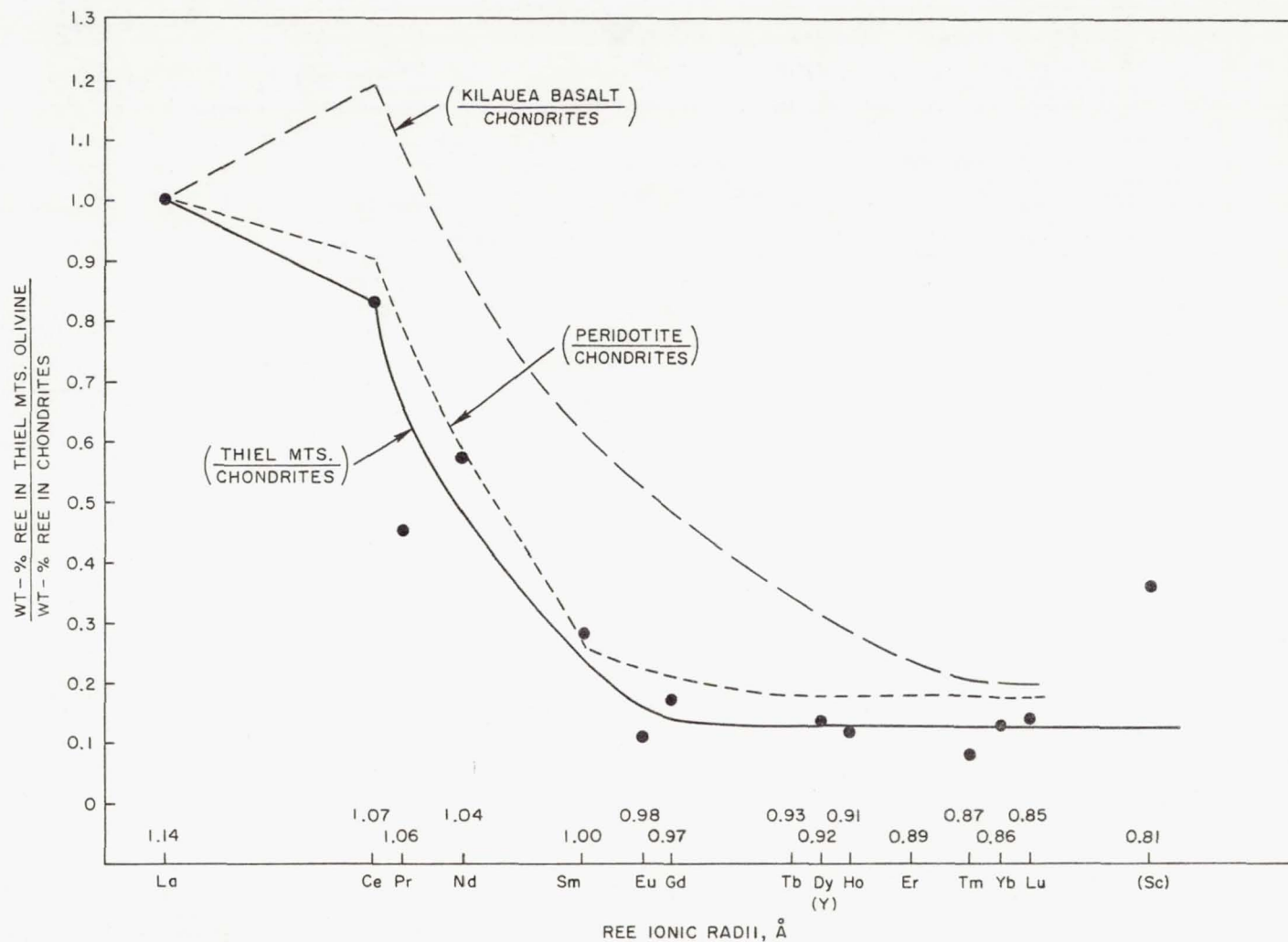


Fig. 5--Ratios of the normalized weight-% in REE in Thiel Mountains pallasite (olivine phase) to the average normalized weight-% REE in 17 chondrites as a function of ionic radii in Å units of trivalent REE. For comparison, the corresponding ratios for terrestrial Kilauea Iki-22 basalt and Wesselton Mine peridotite are included (see REE-1)

compared to a factor of ~ 1.8 in the olivine phase in Brenham. The terrestrial REE distributions of a basalt in combination with a samarskite mineral might conceivably show a composite REE distribution as observed in Brenham. With a reduced ionic radius of 0.89 \AA (AHRENS, 1952) for Eu^{+2} , some Eu^{+2} might be retained by diadochic replacement in the ferromagnesium olivine phase.

In a discussion of the differentiation of chondrites and the formation of irons, stony irons, and achondrites, RINGWOOD (1961) suggests that five parts of pallasites are complementary to one part of eucrite, with mesosiderites resulting from mixing or incomplete differentiation from eucritic, pallasitic, and parental chondritic matter. In short, conservation of REE, Y, and Sc abundances in these four different meteorite types does not adequately support the Ringwood suggestion; see SCHMITT (1962) for details.

FRACTIONATION OF REE, Y, AND Sc IN TERRESTRIAL MATTER

Relative to the REE, Y, and Sc in chondrites, fractionation of these elements has occurred (see Fig. 6) in a hornblende eclogitic inclusion from a basic breccia pipe near Delegate, N. S. W., Australia. LOVERING (1962) states that this specimen is an unaltered sample of basic upper-mantle material, and the Rb-Sr data suggests an age of $4.3 \pm 0.2 \times 10^9$ years for it. The absolute abundances of the REE contents are roughly similar (see Table 4 and Fig. 6) for La, Lu, and Sc in the South African and Australian eclogite. According to LOVERING (1962a) the trace-element contents of the South African eclogite may be dubious because of possible contamination by basalt. However, from a consideration of the absolute abundances and distributions of the REE in oceanic basalts (see dashed curve of Fig. 2), it seems improbable that the African eclogite has been seriously contaminated by REE from basalts. Also, note that Ce has differentiated as expected, which would indicate that there was no special oxidation history for these eclogites compared with chondrites.

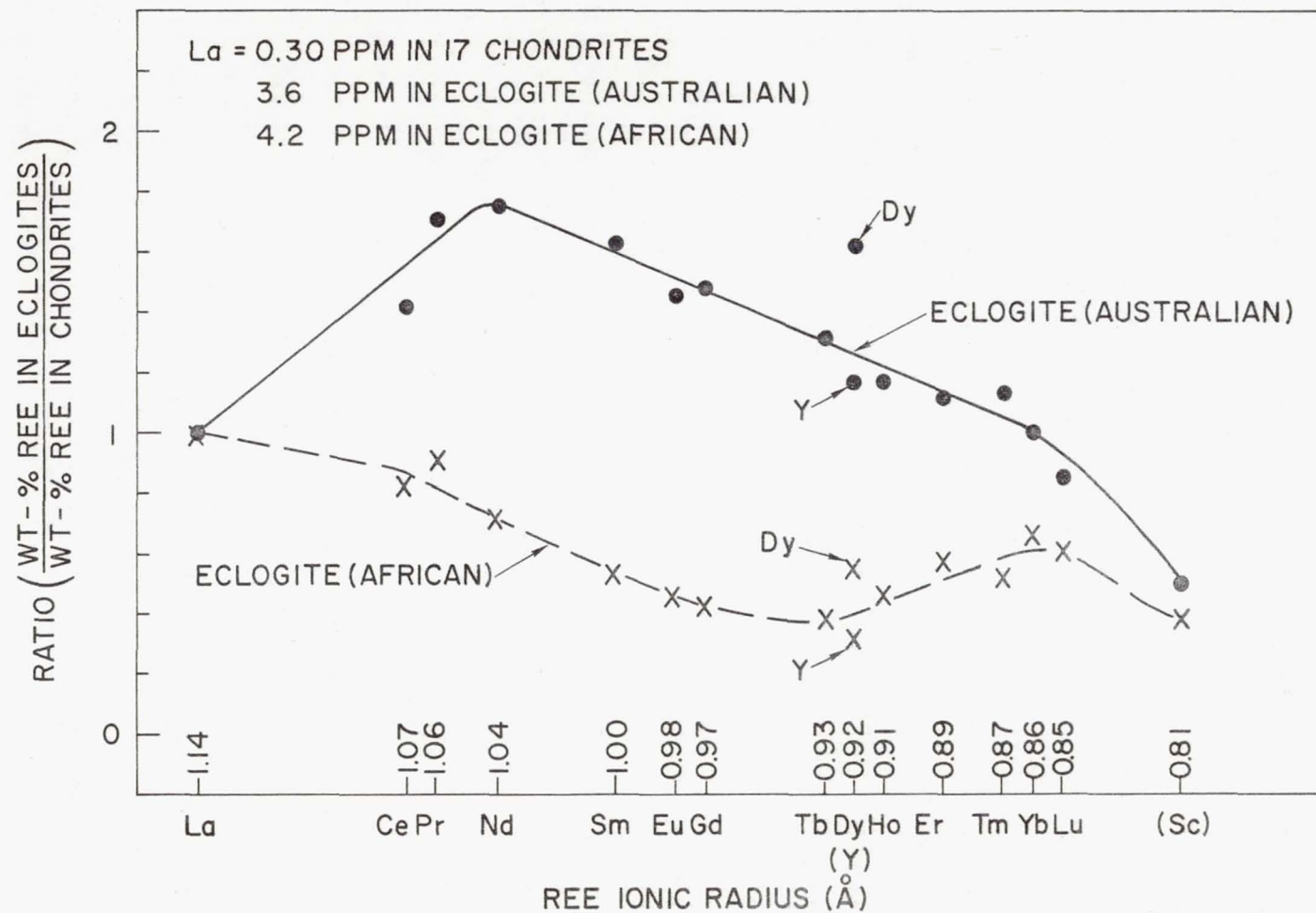


Fig. 6--Ratios of the normalized weight percent of the REE in an Australian eclogite and South African eclogite to those in 17 chondrites as a function of trivalent ionic radius

Table 4

ABSOLUTE AND NORMALIZED ABUNDANCES OF REE, Y, AND Sc IN AN AUSTRALIAN ECLOGITE AND THOLEIITIC BASALT (COLUMBIA PLATEAU) AS DETERMINED BY NEUTRON-ACTIVATION ANALYSIS

Isotope Measured	Radio-nuclide	Absolute Abundances in 10^{-6} g/g Sample (ppm)		Normalized Abundances	
		Eclogite	Basalt	Eclogite	Basalt
La ¹³⁹	40-h La ¹⁴⁰	3.57±0.07	25.3±0.5	1.00	1.00
Ce ¹⁴⁰	32-d Ce ¹⁴¹	14.8±0.2	64.0±1.0	3.95	2.60
Ce ¹⁴²	33-h Ce ¹⁴³	13.4±0.3	67.3±1.4		
Pr ¹⁴¹	19-h Pr ¹⁴²	2.51±0.06	7.90±0.16	0.70	0.31
Nd ¹⁴⁶	11.1-d Nd ¹⁴⁷	11.7±0.2	36.6±0.6	3.28	1.45
Sm ¹⁵²	47-h Sm ¹⁵³	3.84±0.03	10.0±0.2	1.08	0.40
Eu ¹⁵¹	9.3-h Eu ^{152m}	1.24±0.02	2.32±0.03	0.35	0.092
Eu ¹⁵¹	13-y Eu ¹⁵²	1.26±0.04	2.28±0.05		
Gd ¹⁵⁸	18-h Gd ¹⁵⁹	5.17±0.09	10.2±0.2	1.45	0.40
Tb ¹⁵⁹	72-d Tb ¹⁶⁰	0.75±0.01	1.66±0.02	0.21	0.066
Dy ¹⁶⁴	2.3-h Dy ¹⁶⁵	5.7±0.5	9.6±0.2	1.6	0.38
Ho ¹⁶⁵	27-h Ho ¹⁶⁶	1.00±0.01	1.94±0.04	0.28	0.077
Er ¹⁶⁸	9.4-d Er ¹⁶⁹	2.62±0.10	5.60±0.24	0.77	0.214
Er ¹⁷⁰	7.5-h Er ¹⁷¹	2.88±0.03	5.25±0.08		
Tm ¹⁶⁹	129-d Tm ¹⁷⁰	0.41±0.02	0.75±0.02	0.115	0.030
Yb ¹⁶⁸	32-d Yb ¹⁶⁹	1.95±0.06	3.88±0.06	0.57	
Yb ¹⁷⁴	4.2-d Yb ¹⁷⁵	2.05±0.03			
Lu ¹⁷⁶	6.8-d Lu ¹⁷⁷	0.32±0.01	0.64±0.01	0.088	0.0253
Y ⁸⁹	64-h Y ⁹⁰	27±0.4	53±3.0	7.6	2.08
Sc ⁴⁵	85-d Sc ⁴⁶	48±1	37±0.8	13.6	1.46

The REE distribution for the earth's crust may be best represented by that found in 7 different sediments (see HASKIN and GEHL, 1962 and SCHMITT, et al., 1963c). Assuming an initial chondritic distribution for the earth for the non-volatile elements and also assuming that there was no appreciable mixing in the mantle, the REE distribution in the upper mantle will be uniformly enriched in the heavy REE by a factor of ~ 5 and depleted monotonically in the light REE. Neither of the eclogitic specimens represent such a distribution. Within the framework of the assumptions given above, neither of these two eclogitic specimens represent unfractonated upper-mantle matter.

Fractionation (Fig. 7) of the REE, Y, and Sc in the Columbia Plateau basalt appears to be similar to that found in Kilauea Iki-22 basalt (see REE-I). The severity of differentiation for the REE and Y in the Kilauea basalt exceeds that in the Columbia Plateau basalt, whereas nearly the same degree of differentiation has occurred in these basalts for Sc. If the curves from Gd to Ce are extrapolated back to La, it appears that La has been fractionated more in Kilauea than in Columbia plateau basalt. The general similarity in REE and Y fractionation in two basalts compared to that observed in two nakhlitic achondritic meteorites (see Fig. 2) strongly suggests that the nakhlites originated from terrestrial-like volcanic action.

PROPOSED SHELL MODEL FOR PARENT METEORITIC BODY

ANDRES and Gales (1961) have recently reviewed and summarized the three principal theories on the origin of meteorites, i. e., in planetary sized bodies (LOVERING, 1958, and RINGWOOD, 1961), primary and secondary objects (UREY, 1956, 1959), and in asteroidal-sized bodies (FISH, et al., 1960).

The following shell model (SCHMITT, 1963) for a single parent meteoritic body after FISH, et al. (1960), and RINGWOOD (1961) satisfies the abundance data for the lengthy and sensitive group of chemical elements, the REE, Y, and Sc, obtained to date in the entire spectrum of meteorites

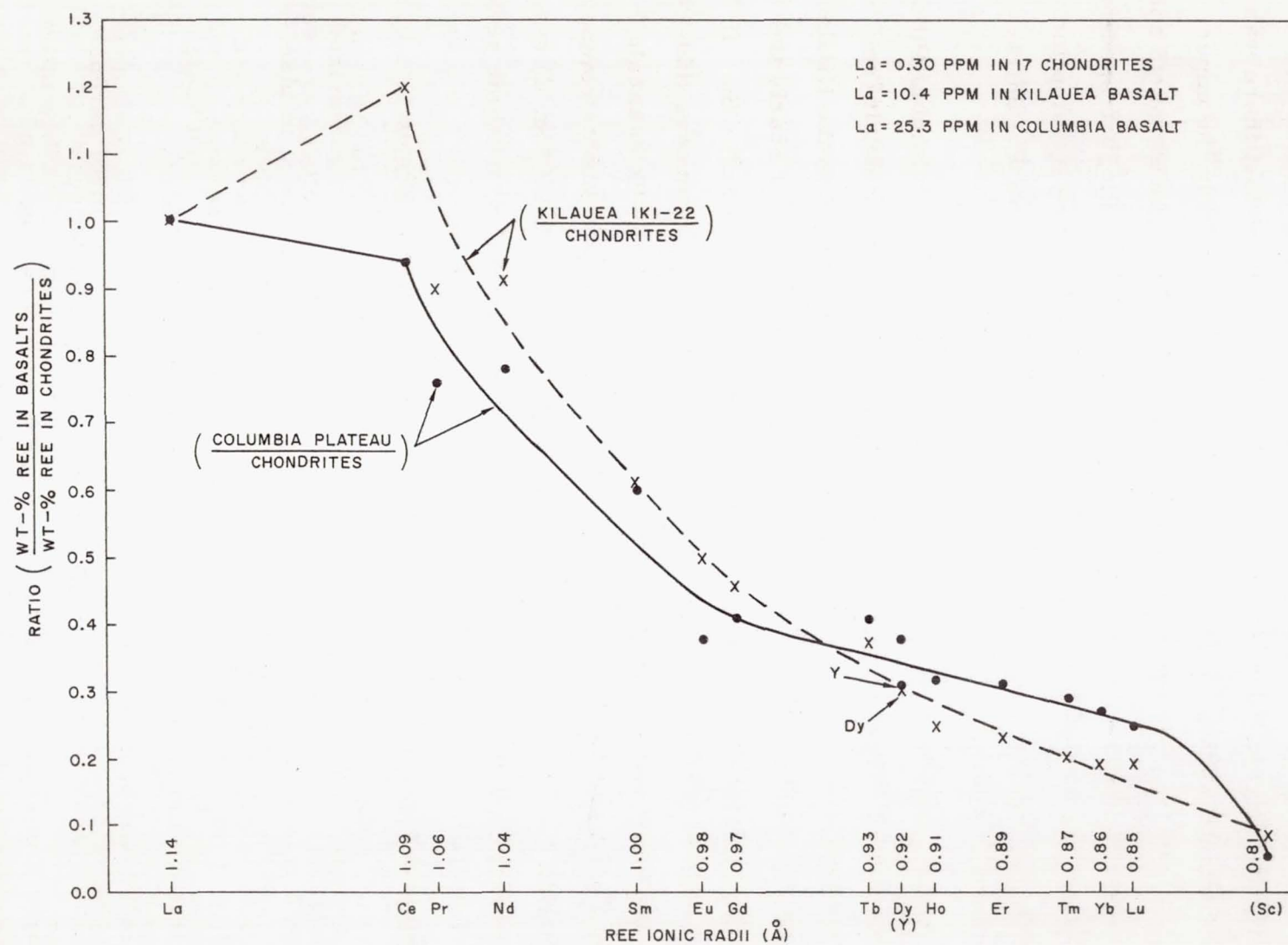


Fig. 7--Ratios of normalized weight percent of the REE, Y, and Sc in Columbia Plateau tholeiitic basalt and in Kilauea Iki-22 basalt to those in 17 chondrites as a function of trivalent ionic radius

(in 32 meteorites)--i. e., in all chondritic categories, including the various types of carbonaceous chondrites, in Ca-poor (chondritic-like) achondrites, in Ca-rich achondrites, in mesosiderites, in pallasites, and finally in iron meteorites, as reported here and in REE-I. Only the pallasitic and iron meteorites were "finds," all the others analyzed being "falls." The observed nonfractionation of the REE and Y in all the chondrites (Type I carbonaceous chondrites excluded because of possible REE fractionation relative to Y) and in Ca-rich achondrites, which account for over 90% of the observed falls, suggests that the chemical history of the parent body subsequent to accretion may have been uncomplicated.

Let us assume the validity of WOOD's (1962) hypothesis concerning the accretion of primary matter expelled by the sun at high temperatures. If rare gases are trapped by the condensation of high-temperature minerals, the highly lithophilic and nonvolatile REE are most certainly trapped and perhaps rather uniformly distributed throughout the small chondrules if the cooling process were rapid enough. An accretion of dust and chondrules into a planetesimal or small planet-sized body is followed by segregation into an Fe-Ni core and outer silicate layers as a result of heating, either by short-lived radionuclides, viz. Al^{26} , or by long-lived radioactivities, or by combinations of these two, depending on the size of the parent body.

The REE, Y, and Sc data for chondrites and achondrites are satisfied by postulating successive shells or layers, as depicted in Fig. 8. In this figure the relative weights in the various shells are equated to the observed fall distribution of the various types of meteorites. The authors are well aware of the pitfalls of such an assumption--e. g., various degrees of friability of meteorites. The shells have resulted from the melting and recrystallization of "primitive" chondritic matter. Obviously, any original chondrules will be destroyed during the melting process with subsequent formation in the chondritic shell, as proposed by RINGWOOD (1961) in the cavitation process. The carbonaceous chondrites assumed to constitute the surface remain unchanged.

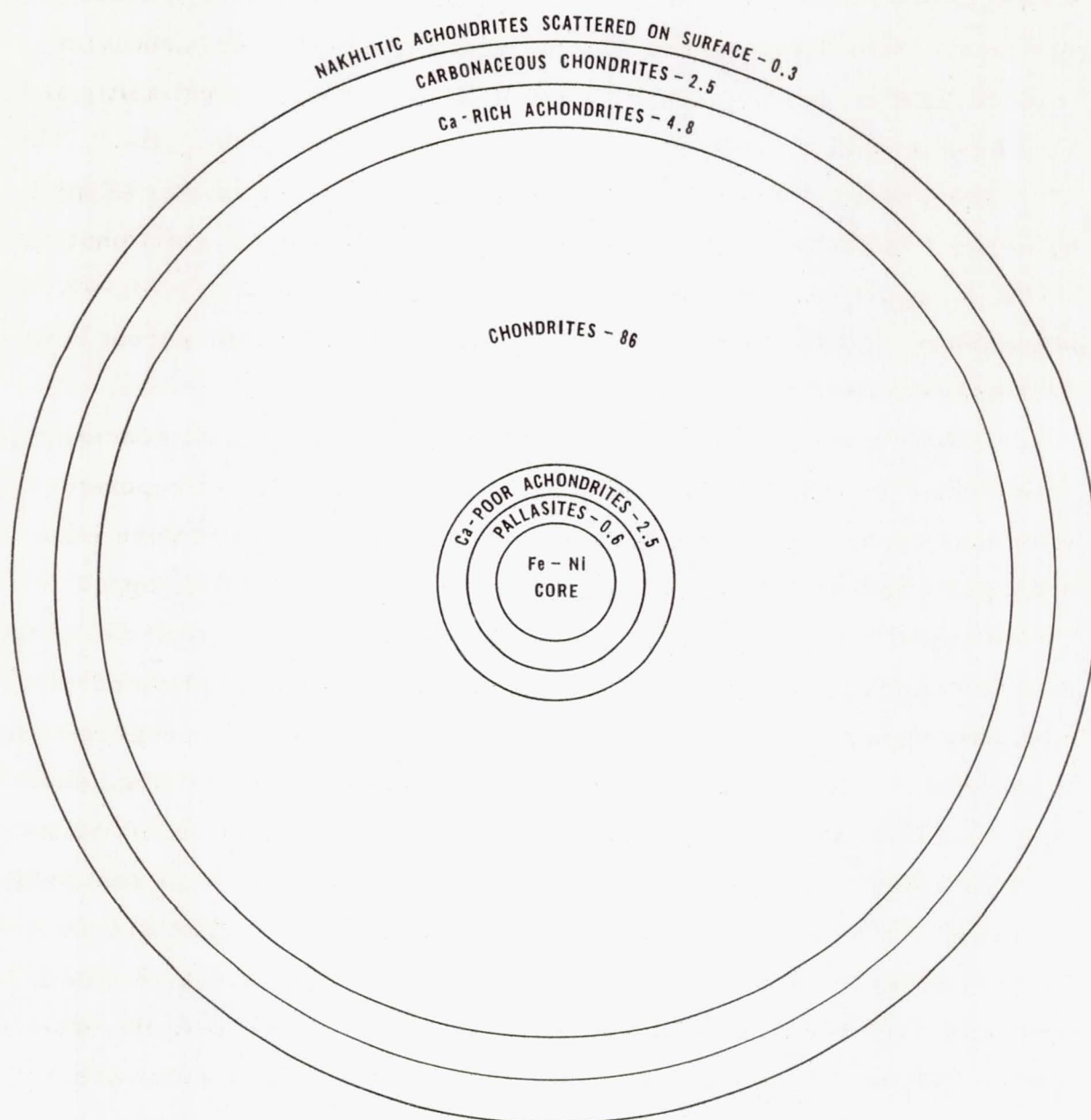


Fig. 8--Proposed meteoritic parent body that satisfies the abundance data of the 14 REE, Sc, and Y for 32 different meteorites (17 chondrites, 2 pallasites, 3 Ca-poor achondrites, 2 mesosiderites, 4 Ca-rich achondrites, 2 nakhlitic achondrites, and 2 iron meteorites. Numbers indicate relative observed falls in percentage $\times 100$ (relative volumes, assumed to be proportional to observed falls, have not been drawn to scale). Fe-FeS eutectic zone and also any water zone has been left out; these probably occur just immediately above the core and under the carbonaceous chondritic zone, respectively

The existence of low REE, Y, and Sc contents in pallasites is consistent with unfavorable replacement of REE and Y in olivine minerals. The small observed content of REE in iron meteorites (ratio of REE in irons to REE in chondrites $\sim 10^{-4}$) simply means that the REE minerals have been efficiently swept out of a molten core (FISH, et al., 1960).

Upon heating of the primitive chondritic matter into a magma followed by crystallization, a calculated fraction (see below), ~ 0.35 of the total REE and Y in the original primitive matter, has been concentrated in the Ca-rich achondrites. On the basis of observed falls, 86% and 4.8% of all meteorites are chondrites and Ca-rich achondrites, respectively. The bronzites and hypersthene constitute $\sim 96\%$ of the chondrites, and the enstatites represent only a small fraction ($\sim 1\%$) of the total. Also, on a volatile free basis, water and carbon excluded, the abundances of La (a typical REE) are: 0.25 ± 0.01 ppm in 2 Type I carbonaceous chondrites, 0.40 ± 0.06 in 2 Type II and 0.40 ± 0.06 in 2 carbonaceous pigeonitic chondrites, 0.33 ± 0.03 ppm in 7 bronzitic and hypersthene chondrites, and 0.20 ± 0.05 in 3 enstatitic chondrites. Also, the La abundance in 4 eucritic and howarditic achondrites (which constitute $\sim 93\%$ of the Ca-rich achondrites) is 3.7 ± 0.8 ppm. All the \pm values given above represent the mean deviations. The Ca-rich achondrites are presumed to be derived from differentiated primitive matter (see MASON, 1962a; MOORE, 1962; and LOVERING, 1962).

In order to calculate the fraction of the REE (La representative) that have migrated into the Ca-rich achondrites from the primitive achondritic matter, let x ppm = La concentration in the original undifferentiated 86 volumes of primitive chondritic matter and in the original 4.8 volumes of Ca-rich achondrites. 0.33 ppm = La concentration in 86 differentiated bronzitic and hypersthene matter. 3.7 ppm = La concentration in 4.8 volumes of Ca-rich achondrites. y = fraction of La lost to Ca-rich achondrites. The loss and gain of La may be simply stated in the equations

$$x - xy = 0.33 ,$$

$$x + 18 xy = 3.7 .$$

The values of x and y are $x = 0.51$ ppm La and $y = 0.35$ ppm La.

A calculated La abundance of 0.51 ppm for the primitive meteoritic matter disagrees violently with the La abundances of 0.25 ± 0.01 ppm in two Type I carbonaceous chondrites. The calculated value of 0.51 ppm is also $\sim 25\%$ higher than the mean value of 0.40 ± 0.06 in Type III carbonaceous pigeonitic chondrites, but the 0.51 ppm agrees within experimental error with two of the four La abundances in Type II and Type III carbonaceous chondrites. Any Type II and III carbonaceous chondrites may be placed as the surface layer of the shell model.

This shell model is certainly incompatible with Type I carbonaceous chondrites for primitive matter. As pointed out in a prior observation, the differences in the Ce/La ratios for Orgueil and Ivuna indicates dissimilarity in their constituencies, thereby relegating Orgueil and Ivuna to secondary and not primitive matter. Differences in the concentrations of Ca and P in Orgueil and Ivuna (WIIK, 1956) also suggest a secondary stage for Type I carbonaceous chondrites.

The similarity in La abundances of 0.25 ± 0.01 ppm (on an H_2O - and C-free basis) in 2 Type I carbonaceous and 0.20 ± 0.05 ppm in 3 enstatitic chondrites suggests a common history and another parent meteoritic body for these two chondritic classes. High abundances of chalcophilic elements like Cd (SCHMITT, 1963b) and of primordial gases (SIGNER and SUESS, 1963) in both enstatitic and carbonaceous chondrites also links these chondrites to some common history, apart from the ordinary chondrites.

With 8.7 ± 0.5 ppm of Sc in 5 bronzitic and hypersthene chondrites, a similar calculation for Sc yields a primitive Sc abundance of 10.2 ppm and 0.15 for the fraction of Sc that migrated into the Ca-rich achondritic layer. The abundance of 10.2 ppm for Sc compares with averages of 10.2 ± 0.5 ppm in 2 Type II carbonaceous chondrites, 11.7 ± 1.5 ppm in 2 carbonaceous pigeonitic (or Type III carbonaceous) chondrites, 7.3 ± 0.4 ppm in 2 Type I carbonaceous chondrites, and 6.3 ± 1.0 ppm in enstatitic chondrites. The hiatus between calculated and observed abundances of

Sc in "primitive" and Type I carbonaceous chondrites and the relatively close Sc agreement between Type I carbonaceous and enstatitic chondrites suggests the same conclusions reached with calculated and observed REE data, La being representative.

Calculations and conclusions for Y are similar to those for La and the REE. With 2.1 ± 0.1 ppm Y in 4 bronzitic and hypersthene chondrites, a calculated primitive Y abundance of 3.2 ppm compares with 2.1 ppm in Type II carbonaceous chondrites, 2.5 ± 0.1 ppm in 2 carbonaceous pigeonitic (or Type III carbonaceous) chondrites, 2.1 ± 0.2 ppm in 2 Type I carbonaceous chondrites, and 1.2 ± 0.3 ppm in 2 enstatitic chondrites.

Other trace lithophiles and nonvolatile elements such as Ba, U, and Th (see REE-I for references) are enriched by a factor of ~ 10 to 12 times (as are the REE) in Ca-rich achondrites compared with ordinary chondrites; consequently, the element Ca is not chiefly responsible for REE enrichment in the Ca-rich achondrites. In general, the large radii of these heavy elements prevent isomorphous replacement in the crystal lattices of minerals such as olivines and pyroxenes.

In this shell model, an analogy of a chondritic earth model has been used. Namely, with the original La content of the earth assumed at 0.30 ppm (average in 17 chondrites) and the assumed (HASKIN and GEHL, 1962; and SCHMITT, *et al.*, 1963c) crustal La content at ~ 10 ppm, the enrichment factor for REE varies from ~ 30 for La to ~ 6 for Gd and ~ 6 from Gd to Lu. The slower cooling of the earth's crust compared with that of a smaller chondritic body may have been responsible for the observed REE and Y fractionation in the earth's crust relative to the chondritic REE and Y distribution. Absence of REE and Y fractionation in Ca-rich achondrites may suggest a faster cooling rate for this layer.

In a melting and crystallization concentration process, the REE and Y contents increase with increasing radius; in other words, that part of the chondritic shell closest to the Fe-Ni core has lost relatively more REE and Y. A positive concentration gradient of REE and Y content

exists toward the surface. The main shell of bronzitic and hypersthene chondrites rests atop the Ca-poor achondritic layer. An Fe-FeS eutectic zone has been neglected, since the REE have not been observed to concentrate in troilite. Type II carbonaceous and pigeonitic chondrites may constitute the carbonaceous layer. If the enstatitic chondrites were relegated to the bottom of the chondritic shell because of the low La/Si ratio of 0.24 ± 0.04 , serious difficulty would be encountered because of the high chalcophilic elemental (SCHMITT, 1963b) and primordial gas abundances (SIGNER and SUESS, 1963).

Ca-poor achondrites have been relegated to the shell above the pallasitic layer. REE and Y contents in these achondrites range from ~ 0.05 to 0.6 times the REE and Y contents in chondrites; moreover, in all cases, the REE, Y, and Sc have been severely fractionated relative to chondrites (see Fig. 1) and in somewhat similar patterns. Actually, the Ca content and mineralogy of these achondrites are quite similar to ordinary chondrites (UREY and CRAIG, 1953). On an absolute basis, the content of Sc remained essentially unchanged in these Ca-poor achondrites compared with chondrites. Simple conservation of Sc shows that ~ 0.15 of the original Sc in the primitive chondritic layer migrated to the Ca-rich achondritic zone compared with ~ 0.35 for the REE and Y. This is consistent with the observed (GOLDSCHMIDT, 1954) enhancement of Sc content in Mg and Fe minerals relative to REE and Y. If Sc were isomorphously substituted (owing to its smaller ionic radius) in Mg and Fe minerals in chondrules, removal of the Sc would necessitate high temperatures.

Low abundances of REE and Y in two of the three Ca-poor achondrites analyzed may result from the lack of isomorphous replacement of the REE and Y in the olivine and enstatitic phases, which constitute appreciable fractions of these meteorites. In Norton County, a Ca-poor achondrite, the absolute and normalized abundances of the heavy REE, Gd to Lu, were indistinguishable compared with chondrites, whereas the light REE, La to Sm, have been fractionated (depleted) monotonically to a La content

~ 0.5 times compared with chondrites; also, Eu was depleted by a factor of 4.2, indicating an appreciable reducing environment for $\text{Eu}^{+3} \rightarrow \text{Eu}^{+2}$. From a microprobe analysis of some minerals in Norton County, KEIL and FREDRIKSSON (1963) have also concluded that Norton County was formed in a strongly reducing environment. In the pallasite Brenham, Eu was depleted by a factor of ~ 1.8 , indicating appreciable Eu reduction, followed by possible incorporation of Eu^{+2} in minerals not present in the silicate phase.

The mesosideritic layer has not been included in the shell model for a number of reasons. First, the REE and Y contents have not been fractionated in Estherville and have fractionated, with Eu enrichment, in Veramin, relative to chondrites. Second, absolute abundances of the REE and Y in the silicate phase of Estherville are ~ 4 times greater than in the chondrites and those in Veramin are about the same as in the chondrites. Relative to the chondrites, Sc has been enriched ~ 2 times in both Estherville and Veramin. An enrichment of the REE and Y is consistent only with a layer outside the main chondritic layer (for Estherville), and REE and Y fractionation is consistent with a layer under the main chondritic layer. Such an inconsistency, or irreproducibility, for the REE, Y, and Sc abundances relegates the mesosiderites to a nonstationary layer, i.e., formation of mesosiderites probably involves a rapid and nonequilibrium physiochemical process.

Finally, the nakhlitic meteorites, which account for only $\sim 3\%$ of observed meteorite falls, may be located on the surface of the parent body. Briefly, since the REE and Y have been fractionated identically in both Nakhla and Lafayette and since the fractionation is very similar to terrestrial basalt (Kilauea Iki-22 and Columbia Plateau), volcanic action may be postulated for the fractionation phenomenon (SCHMITT and SMITH, 1963). In the analysis of the meteorite Nakhla, PRIOR (1912) reported the resemblance of Nakhla to a fine-grained basalt, and MASON (1963) states that the two meteorites Nakhla and Lafayette are mineralogically

indistinguishable. Furthermore, it is suggested that the volcanic root resides in the Ca-rich achondritic layer. On an absolute basis, the La content of these nakhlitic meteorites is ~ 0.5 times that present in Ca-rich achondrites. Assuming that the volume ratio of nakhlites to all Ca-rich achondrites is $0.3/4.8$, or 0.06 (which is very poor statistics), about 3% to 6% of the Ca-rich achondritic volume should be enriched in the heavy REE and Y. With only 37 observed Ca-rich achondritic falls, only ~ 2 of these achondrites should show an enrichment factor. Only a small fraction of the surface would be covered with this basalt; the surface layer of the parent meteoritic body would therefore still remain carbonaceous chondritic in composition.

FISH, et al., (1960) suggest a shell arrangement with decreasing radius of carbonaceous chondrites, friable chondrites, chondrites, mesosiderites, achondrites, pallasites, and finally, an iron-nickel core. Their model would suggest no significant change in the La/Si atomic ratio for the carbonaceous, friable, and chondritic layers, which is contrary to observation. On the other hand, the REE, Y, and Sc data may be partially satisfied by the model proposed by FISH, et al., if the Ca-rich achondrites are situated immediately below the sintered chondritic shell. This latter arrangement would prevent the normal ascent of a lighter basalt.

It should be emphasized that the present shell model satisfies the REE, Y, and Sc abundances determined in all meteoritic phases except for the special Type I carbonaceous and enstatitic chondrites. Any other proposed model must also be consistent with the abundance data of this long series of very chemically sensitive REE, Y, and Sc elements.

Abundances of other trace, lithophilic, and nonvolatile elements have usually been determined in only a few types of meteorites, chondrites, and Ca-rich achondrites. For the elements Ba and U (REED, et al., 1960), and Sr (GAST, 1960), the shell model is consistent with their observed fractionations.

Some of the abundances that define the "cosmic" abundance distribution

curve will certainly be affected by the inclusion of Ca-rich achondrites with ordinary chondrites in a single parent meteoritic body. In general, the abundances of trace, lithophilic, nonvolatile, and heavy elements found in ordinary bronzitic and hypersthene chondrites must be increased by $\sim 35\%$ in order to calculate the primitive abundances. Corrections must be applied to elements that are not readily accommodated in olivine or enstatitic minerals, and that crystallize in the residual minerals of a chondritic magma. In general, elements with ionic radii $> 0.9 \text{ \AA}$ satisfy these criteria.

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